

# Interplane magnetic coupling effects in the multilattice compound $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$

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We investigate the interplane magnetic coupling of the multilattice compound  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  by means of a bilayer Hubbard model with inequivalent planes. We evaluate the spin response, effective interaction and the intra- and interplane spin-spin relaxation times within the fluctuation exchange approximation. We show that strong in-plane antiferromagnetic fluctuations are responsible for a magnetic coupling between the planes, which in turns leads to a tendency of the fluctuation in the two planes to equalize. This equalization effect grows with increasing in-plane antiferromagnetic fluctuations, i. e., with decreasing temperature and decreasing doping, while it is completely absent when the in-layer correlation length becomes of the order of one lattice spacing. Our results provide a good qualitative description of NMR and NQR experiments in  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ .

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Although many models for high-T<sub>c</sub> cuprates are restricted to a single layer, it has become clear that both superconducting and magnetic properties of these materials are affected by the coupling between two or more layers. A rather strong coupling between the layers has been observed principally by inelastic neutron scattering<sup>1</sup> (INS) and nuclear magnetic resonance<sup>2-5</sup> (NMR). Furthermore, the observation of a qualitatively different behavior of the odd and even channel in INS<sup>6</sup> and of a bilayer splitting of the Fermi surface found in angular resolved photoemission experiments (ARPES)<sup>7,8</sup> demonstrate that low energy excitations of cuprates are affected by the presence of more than one layer per unit cell. An exciting perspective on the nature of the coupling between CuO<sub>2</sub>-layers was offered by NMR experiments by Stern *et al.* on  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  (247). This material has a variety of structural similarities to the extensively studied  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (123) and  $\text{YBa}_2\text{Cu}_4\text{O}_8$  (124) systems. The compound 247 can be considered as a natural multilattice, whose bilayers are build up of one CuO<sub>2</sub> layer which belongs to the 123 block and one layer to the 124 block. Based on the analysis of the NQR spectra it turned out that the charge carrier content in these nonequivalent adjacent layers is very close to that of the related parent compounds of the two blocks, 123 and 247. Interestingly, the highest transition temperature ( $T_c = 95\text{ K}$ ) occurs in the 247 compound, in comparison with the 92 K of 123 and 82 K of the 124 system.

In this paper, we want to provide a theoretical understanding in terms of a microscopic model of some striking experimental observations of Refs. 2,3, namely: (i) the spin-spin relaxation rates  $T_{2G}^{-1}$  of the two layers in  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ , measured in a spin-echo double resonance experiment, behave very similarly as a function of temperature, despite the different doping of the layers; (ii) the spin-spin relaxation rate in the 124 (247) layer of  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  is reduced (enhanced) with respect to one of the constituent compound at low temperatures; (iii) the interplane transverse relaxation rate, increases for

decreasing temperature faster than the intraplane one. The overall features that can be inferred from these experiments are that, for high temperatures the magnetic fluctuations in the two layers of  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  are disconnected and each layer behaves similarly to the corresponding parent compounds, whereas for decreasing temperatures, the increasing interlayer magnetic coupling enforces even the slightly overdoped plane to behave like an underdoped system and vice-versa.

To describe the strong electronic correlations in the system  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ , consisting of two layers with different charge carrier concentration, we extend the standard single-band Hubbard model to include two layers coupled by an interplane hopping matrix element  $t_{\perp}$ . Furthermore, to produce a different hole concentration in the planes, we introduce an on-site energy  $\delta$  in the second plane. After Fourier transformation of the kinetic part the Hamiltonian reads:

$$H = \sum_{\substack{l_1, l_2 \\ \mathbf{k}, \sigma}} [H_o(\mathbf{k})]_{l_1, l_2} \cdot c_{\mathbf{k}, l_1, \sigma}^{\dagger} c_{\mathbf{k}, l_2, \sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} . \quad (1)$$

where  $c_{\mathbf{k}, l_1, \sigma}^{\dagger}$  creates a particle with spin  $\sigma$  and momentum  $\mathbf{k}$  in layer  $l_1$ , and  $i$  runs over the sites of the two layers. In contrast to the standard notation in the monolayer case,  $\epsilon_{\mathbf{k}}$  is replaced by the  $2 \times 2$  matrix.  $H_o(\mathbf{k})$ , whose components are  $[H_o(\mathbf{k})]_{11} = \epsilon_{\mathbf{k}} - \mu$ ,  $[H_o(\mathbf{k})]_{22} = \epsilon_{\mathbf{k}} + \delta - \mu$  and  $[H_o(\mathbf{k})]_{12} = [H_o(\mathbf{k})]_{21} = t_{\perp}$ . The bare dispersion  $\epsilon_{\mathbf{k}}$  includes second ( $t'$ ) and third ( $t''$ ) nearest-neighbor hopping processes to better model the Fermi surface of the cuprates (see, e.g.<sup>9,10</sup>). The properties of the interacting system are deduced from the Green's function and form the dynamic two-particle susceptibilities in the framework of the fluctuation exchange approximation (FLEX)<sup>11</sup>. In this approximation, bubble and ladder diagrams are summed up in infinite order and the resulting coupled set of equations is solved self-consistently. Although this approximation is suitable to describe magnetic fluctuations and band shadow

features,<sup>12</sup> it fails to reproduce the pseudogap behavior in the spectral function and in the magnetic excitations at low temperatures and low doping<sup>13</sup>, probably due to the lack of conservation at the two-particle level and to the neglection of interference effects between particle-particle and particle-hole channel. Moreover, on the quantitative level, antiferromagnetic fluctuations appear to be underestimated<sup>14</sup> with respect to exact results.

In order to understand the systematics and the parameter dependence of the interplane magnetic coupling, we first focus our attention on two systems with different parameter sets  $t'/t, t''/t$  corresponding to strong and weak antiferromagnetic fluctuations. Specifically, we introduce a first system, for simplicity labeled by “A”, with  $t' = -0.38t, t'' = -0.06t$ , and a second one (“B”) with  $t' = -0.20t, t'' = 0.15t$ . System A shows much stronger antiferromagnetic fluctuations due to the underlying Fermi surface in comparison to B. The Hubbard interaction takes an intermediate value  $U = 4t$ , as appropriate for a perturbative calculation, and  $t_{\perp} = 0.4t$  (cf.<sup>15,16,14</sup>). Within the self-consistency cycle, we fix the on-site energy  $\delta$  and the particle number  $n_1 = 1 - x_1$  of the first layer, while the chemical potential  $\mu$  and the particle number of the second plane  $n_2 = 1 - x_2$  are adjusted at each step. Keeping this way the doping of the first plane fixed and changing only the doping of the second one, we investigate a possible magnetic coupling between the layers. Our question is whether the magnetic fluctuations of the first plane are influenced by the doping of the second plane and vice-versa.

With this in mind, we consider in Fig. 1 the static spin-spin function  $\chi_{ll}^{zz}(\mathbf{q}, \omega = 0)$  along the standard path  $(0, 0) \rightarrow (\pi, 0) \rightarrow (\pi, \pi) \rightarrow (0, 0)$  in the Brillouin zone for the two layers ( $l = 1$  and  $l = 2$ ) and both systems A and B.

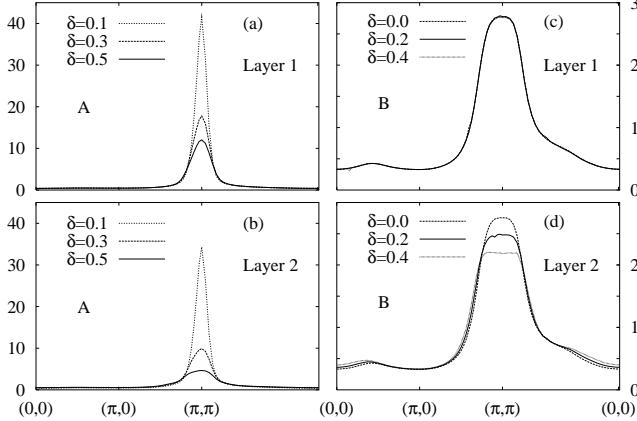


FIG. 1. Static spin susceptibility  $\chi_{ll}^{zz}(\mathbf{q}, \omega = 0)$  along the standard path in the Brillouin zone for the two layers and both systems A and B (see text). (The temperature is  $T = 0.02t$ , and  $x_1 = 0.08$ )

As one can clearly see from Fig. 1(a) and (b) for the case of system A, the spin response in the two planes is strongly peaked at the antiferromagnetic wave vector

$\mathbf{Q} = (\pi, \pi)$  indicating pronounced antiferromagnetic fluctuations in the Hubbard planes. The striking result is that data for a plane with a given doping ( $l_1 = 1$ ) also depend on whether this plane is coupled with an equivalent one or with a more or less doped one, depending of the value of  $\delta$ . This behavior clearly suggests a strong magnetic connection between the planes. The decrease of the spin susceptibility with increasing  $\delta$  [see Fig. 1(a) and (b)] is due to the increased hole concentration of the total system moving it further away from half filling where antiferromagnetism is most pronounced. The situation is rather different for system B, whose data are shown in Fig. 1(c) and (d). Here, a variation of  $\delta$  influences the susceptibility of the second plane only, whereas the first plane, with constant charge carrier concentration, is almost not affected at all. Thus, the magnetic fluctuations in the first plane are disconnected from the fluctuations in the second one. Only for a system with sufficiently strong antiferromagnetic correlations and a correlation length larger than a few lattice constants, the two inequivalent layers turn out to be strongly coupled, as in case A.

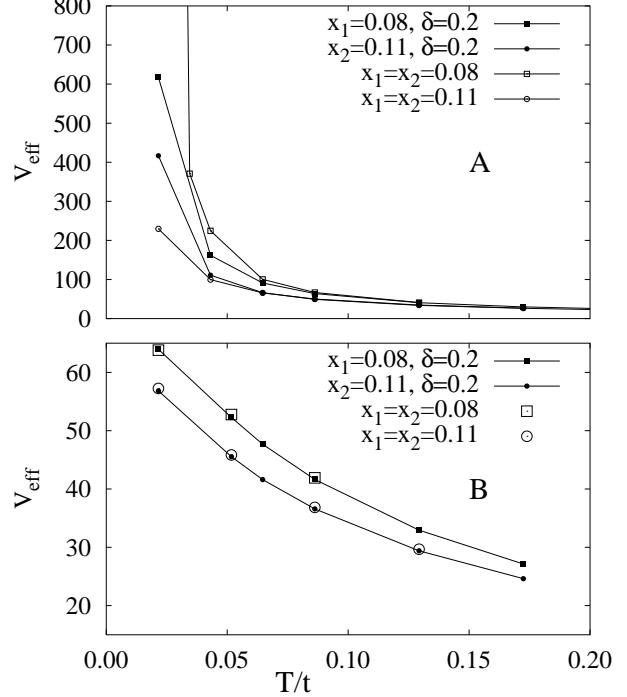


FIG. 2. Effective interaction  $V_{\text{eff}}(\mathbf{Q}, \omega = 0)$  vs.  $T$  for a system with inequivalent layers ( $x_1 = 0.08$  and  $x_2 = 0.11$ ) in comparison with the corresponding bilayer systems with  $x_1 = x_2 = 0.08$  and  $x_1 = x_2 = 0.11$ . The upper panel shows the results for system A and the lower for B.

We now ask to what extent each layer of a system with inequivalent layers compares to the corresponding system with equivalent layers (i.e.,  $\delta = 0$ ). Therefore, we show in Fig. 2 the effective interaction  $V_{\text{eff}}(\mathbf{Q}, \omega = 0)$  at the antiferromagnetic wave vector  $\mathbf{Q} = (\pi, \pi)$  as a function of temperature for systems with inequivalent layers ( $x_1 = 0.08, x_2 = 0.11, \delta = 0.2$ ) in comparison

with the two corresponding systems with identical layers ( $x_1 = x_2 = 0.08$ ) and ( $x_1 = x_2 = 0.11$ ). In the upper panel of Fig. 2 we show the results for system A. Here, we clearly observe considerable differences between each of the layers in the inequivalent-layer system with respect to its counterparts in the equivalent-layer system at low temperatures. Specifically, the effective interactions in the two layers of the system with inequivalent layers are “enclosed” between the ones in the corresponding layers of the system with identical layers. Thus, the interlayer magnetic correlations lead to a tendency for the magnetic fluctuations of the two planes to equalize although their doping is different. The results for system B are strikingly different as can be seen in the lower panel of Fig. 2. Here, the two planes with different dopings in the inequivalent-layer system behave essentially like the correspondingly doped layers in the system with equivalent layers. For this less magnetic system the fluctuations in the two layers are effectively decoupled and do not influence each other. The results displayed in Fig. 2 thus again show that the antiferromagnetic correlations within the layers lead to a partial equalization of the magnetic fluctuations in the two planes.

A similar magnetic equalization effect has been observed by Scalettar et al.<sup>17</sup> in Quantum-Monte-Carlo simulations of an half-filled layer coupled to a doped one. According to these authors, this equalization effect can be explained by perturbation in  $t_{\perp}$  as a competition between virtual hopping processes between the two layers with energy scale  $\propto t_{\perp}/\delta$  and exchange energy  $J \propto t^2/U$ . We believe that our use of the FLEX approximation is essential in order to additionally explain the crossover from the “equalized” regime (A) to the disconnected one (B), which accounts for the qualitative behavior of the experiments on  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ <sup>3</sup> as we will show below. This is due to the feedback effect of the Green’s functions renormalization in the FLEX approximation.

The equalization effect observed in system A qualitatively describes the experimental situation for the inequivalent-layer system  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ . Indeed, NMR experiments by Stern et al.<sup>3</sup> measuring  $1/T_{2G}$  for the two layers of  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  and for the two corresponding systems with equivalent layers,  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{YBa}_2\text{Cu}_4\text{O}_8$ , show a similar equalization tendency as in Fig. 2. To further demonstrate the similarities of our calculations with the experimental results, we also evaluate the nuclear spin-spin relaxation time  $T_{2G}$  for spins within the two planes. The generalized relaxation rate  $T_{2G}^{ll'}^{-1}$  measuring the interaction between a spin in plane  $l$  and one in  $l'$  is related to a weighted sum over the static spin susceptibility  $\chi_{ll'}^{zz}(\mathbf{q}, \omega = 0)$  where the momenta  $\mathbf{q} \cong \mathbf{Q}$  have the strongest weight.<sup>5,18–20</sup> In our calculations, we assume that the hyperfine coupling constants entering the form factor are the same for both planes and use the values given by Barzykin and Pines<sup>21</sup>. Hence possible difference in the relaxation times of the two planes can solely arise from different spin responses in the layers.

In a previous work on  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ , Millis and Monien, who determined the value of the interlayer exchange interaction from the experimental data of Ref.<sup>3</sup>, have considered an alternative point of view and neglected the difference between the spin susceptibilities of the two layers by considering different hyperfine coupling constants<sup>20</sup>. This approach is justified by our results which show for systems with strong antiferromagnetic correlations a pronounced coupling of the magnetic response of the two layers and thus a tendency to equalization of the susceptibilities. This primarily causes a similar temperature dependence of the two in plane susceptibilities. Nevertheless, quantitatively, the magnitudes of the magnetic response of the two layers stays different.

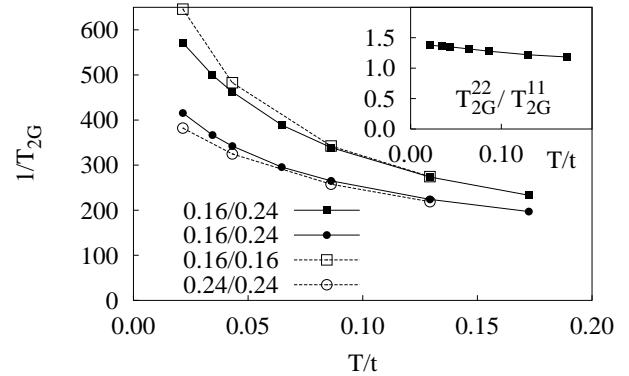


FIG. 3. Temperature dependence of the Gaussian component of the spin-spin relaxation rate  $T_{2G}$  of a bilayer system consisting of two different layers with doping  $x_1 = 0.16$  and  $x_2 = 0.24$ , respectively ( $\delta = 0.4$ ). For comparison, we show also the result for two bilayer systems with equivalent layers with doping  $x_1 = x_2 = 0.16$  and  $x_1 = x_2 = 0.24$ , respectively. The other parameters for all curves are:  $t' = -0.38t$ ,  $t'' = -0.06t$ . The inset shows the ratio  $T_{2G}^{22}/T_{2G}^{11}$  for the case of inequivalent layers.

In Fig. 3 we show the in-plane  $1/T_{2G}$  as a function of temperature for both planes of the system with *inequivalent* layers again in comparison with the data for two corresponding bilayer systems with *equivalent* layers. In this figure, the filled (open) symbols represent the system with inequivalent (equivalent) layers. Furthermore, the squares are related to the data for planes with a doping  $x = 0.16$  and the bullets planes with doping  $x = 0.24$ . Since the main contribution to  $1/T_{2G}$  stems from  $\chi_{ll'}^{zz}(\mathbf{Q}, 0)$ , a system with rather strong magnetism exhibits large relaxation rates  $1/T_{2G}$ . This explains why the layers with  $x = 0.16$  show a smaller relaxation rate compared to  $x = 0.24$ . As expected from the results of Fig. 2, and as observed in NMR experiments at lower temperatures<sup>3</sup>, the heavily doped plane ( $x_2 = 0.24$ ) of the system with inequivalent layers shows stronger magnetic fluctuations than the plane in the corresponding system with equivalent layers ( $x_1 = x_2 = 0.24$ ). Similarly, the magnetism of the lower doped plane ( $x_1 = 0.16$ ) is reduced with respect to the corresponding equivalent-layer system ( $x_1 = x_2 = 0.16$ ) due to the coupling to a more doped plane. As already observed in the results

for the effective interaction shown above, the magnetic fluctuations of the two inequivalent planes with different carrier concentration tend to be partially equalized by interplane coupling effects. The theoretical results in Fig. 3, as well as the experimental results, show a strong increase of  $1/T_{2G}$  with decreasing temperature. However, the experimentally observed decrease of  $1/T_{2G}$  below  $T_{sg} \approx 100$ K due to the opening of a pseudogap in the spin excitation spectrum, is not reproduced by the FLEX approximation, as discussed above.

As NMR experiments show, the spin-lattice relaxation rate  $1/T_{2G}$  has the same temperature dependence in the two planes of  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ <sup>3</sup>, the ratio  $R = (1/T_{2G}^{124})/(1/T_{2G}^{123})$  being temperature independent and approximately  $R \approx 1.4\text{--}1.5$ . This ratio corresponds to  $R = T_{2G}^{22}/T_{2G}^{11}$  in our work, since the 124 layer in the coupled layer structure of  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  is the one with lower doping (here labeled by “1”) and the  $\text{CuO}_2$ -layer from the 123 block corresponds to the second plane in our theoretical study. The theoretical values for  $R$  are shown in the inset of Fig. 3 as a function of temperature. For the parameter set chosen (corresponding to system A),  $R$  is approximately 1.2 – 1.4 and almost temperature-independent, in good agreement with the experimental finding.

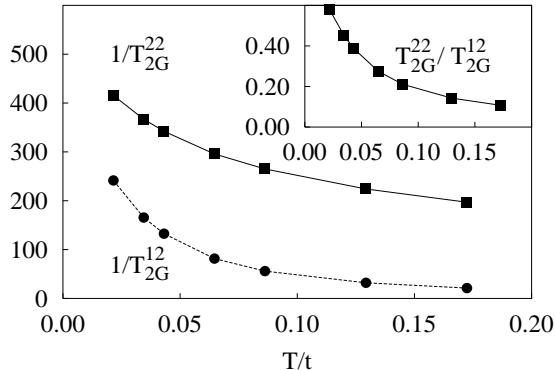


FIG. 4. Intra- and inter-plane spin-spin relaxation rates  $1/T_{2G}^{22}$  and  $1/T_{2G}^{12}$ , respectively, vs.  $T$ . The inset shows the ratio between the two relaxation rates ( $x_1 = 0.16$ ,  $x_2 = 0.24$ ,  $\delta = 0.4$ ).

In order to have a measure of the interplane spin coupling in  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ , Stern *et al.*<sup>4</sup> have carried out NQR spin-echo double-resonance (SEDOR) measurements, as suggested by Monien and Rice<sup>5</sup>. These experiments allow the determination of the inter-layer spin-spin relaxation time  $T_{2G}^{12}$ . The apparent feature in their results is that the inter-plane relaxation rate, although smaller than the in-plane one, increases faster for decreasing temperature, as seen from the temperature dependence of the ratio  $R_{\text{SEDOR}}(T) = T_{2G}^{22}/T_{2G}^{12}$ . For the sake of comparison, we present our theoretical results for the in-plane relaxation rate  $1/T_{2G}^{22}$  together with the inter-plane one  $1/T_{2G}^{12}$  in Fig. 4, the parameters being the same as in Fig. 3. The fact that  $1/T_{2G}^{12}$  grows faster with decreasing temperature is even more apparent in the inset. Here, we

show the temperature dependence of the ratio between the two relaxation rates. Our theoretical results thus reproduce the qualitative behavior observed experimentally. However, for  $T$  smaller than the spin gap the experiments show a saturation effect, which obviously cannot be reproduced within our approximation.

In summary, we have studied the magnetic interplane coupling of the multilattice compound  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$  by modeling it with two inequivalent Hubbard layers coupled by an interlayer hopping  $t_{\perp}$ . If the antiferromagnetic correlation length is less than 1–2 lattice spacings, which happens for high temperatures or for a Fermi surface with suppressed nesting, we find that the two inequivalent layers are disconnected and keep their individual magnetic properties. However, once the antiferromagnetic correlations in the layer with smaller charge carrier concentration is sufficiently large, the single-particle excitations for momenta close to the Fermi surface and, in particular, around the momenta close to  $(\pi, 0)$  as well as the magnetic excitations of the two layers are strongly connected. In this situation, the whole system reacts magnetically as a single system, despite its inhomogeneous charge density. Due to the strong magnetic correlations in the underdoped layer the magnetic in-plane order in the nominally overdoped layer is stabilized via interplane magnetic coupling. Thus, the layer with lower charge carrier concentration acts like an external staggered field. For suitable Fermi surfaces and low enough temperatures, this mechanism makes the magnetic dynamics of both planes indistinguishable. By choosing parameters which lead to a moderate equalization effect, we have qualitatively reproduced the salient features of the in-plane and out-of-plane spin-spin relaxation times observed in NMR measurements on  $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ , as compared with the ones of its constituent compounds.

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